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Seasonal and long term variations of surface ozone concentrations in Malaysian Borneo

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Latif, Mohd Talib; Dominick, Doreena; Ahamad, Fatimah; Ahamad, Nur Shuhada; Khan, Md Firoz; Juneng, Liew; Xiang, Chung Jing; Mohd Nadzir, Mohd Shahrul; Robinson, Andrew D.; Ismail, Marzuki; Mead, Mohammed Iqbal; and Harris, Neil R. P, "Seasonal and long term variations of surface ozone concentrations in Malaysian Borneo" (2016). *Faculty of Science, Medicine and Health - Papers: part A*. 4690.
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Abstract

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Disciplines

Medicine and Health Sciences | Social and Behavioral Sciences

Publication Details

Latif, M., Dominick, D., Ahamad, F., Ahamad, N., Khan, M., Juneng, L., Xiang, C., Mohd Nadzir, M., Robinson, A. D., Ismail, M., Mead, M. & Harris, N. R. P. (2016). Seasonal and long term variations of surface ozone concentrations in Malaysian Borneo. *Science of the Total Environment*, 573 494-504.

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This article was originally published as:

Latif, M., Dominick, D., Ahamad, F., Ahamad, N., Khan, M., Juneng, L., Xiang, C., Mohd Nadzir, M., Robinson, A. D., Ismail, M., Mead, M. & Harris, N. R. P. (2016). Seasonal and long term variations of surface ozone concentrations in Malaysian Borneo. Science of the Total Environment, 573 494-504

Seasonal and long term variations of surface ozone concentrations in Malaysian Borneo

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Abstract

Malaysian Borneo has a lower population density and is an area known for its lush rainforests. However, changes in pollutant profiles are expected due to increasing urbanisation and commercial-industrial activities. This study aims to determine the variation of surface O₃ concentration recorded at seven selected stations in Malaysian Borneo. Hourly surface O₃ data covering the period 2002 to 2013, obtained from the Malaysian Department of Environment (DOE), were analysed using statistical methods. The results show that the concentrations of O₃ recorded in Malaysian Borneo during the study period were below the maximum Malaysian Air Quality Standard of 100 ppbv. The hourly average and maximum O₃ concentrations of 31 and 92 ppbv reported at Bintulu (S3) respectively were the highest among the O₃ concentrations recorded at the sampling stations. Further investigation on O₃ precursors shows that sampling sites located near to local petrochemical industrial activities, such as Bintulu (S3) and Miri (S4), have higher NO₂/NO ratios (between 3.21 and 5.67) compared to other stations. The normalised O₃ values recorded at all stations were higher during the weekend compared to weekdays (unlike its precursors) which suggests the influence of O₃ titration by NO during weekdays. The results also show that there are distinct seasonal variations in O₃ across Borneo. High surface O₃ concentrations were usually observed between August and September at all stations with the exception of station S7 on the east coast. Majority of the stations (except S1 and S6) have recorded increasing averaged maximum concentrations of surface O₃ over the analysed years. Increasing trends of NO₂ and decreasing trends of NO influence the yearly averaged maximum of O₃ especially at S3. This study also shows that variations of meteorological factors such as wind speed and direction, humidity and

temperature influence the concentration of surface O₃.

Keywords: Surface ozone; ozone precursors; meteorological factors; seasonal variations

1. Introduction

Surface ozone (O₃) is a major air pollutant that impacts human health, materials and vegetation (Anenberg et al., 2009; Ishii et al., 2007; Lee et al., 1996). Studies by Levy et al. (2005) and Bell et al. (2014) have shown that O₃ can contribute to higher mortality, especially for elderly people. Several studies, such as West et al. (2006), Jerrett et al. (2009), Guo et al. (2014) and Yang et al. (2014) have indicated that high concentrations of surface O₃ can cause cardiovascular and respiratory dysfunction. The impact of O₃ on human health was found to be further influenced by increasing temperature (Ren et al., 2008). High concentrations of O₃ also damage materials, such as surface coatings and rubber goods, and plants, via membrane damage on leaves therefore effecting the photosynthesis processes (Chaudhary and Agrawal, 2015; Lee et al., 1996). Studies have also shown that surface O₃ can reduce the yield of certain food crops such as paddy-grown rice and grain (Amin, 2014; Debaje, 2014; Ghude et al., 2014; Van Dingenen et al., 2009; Wang and Mauzerall, 2004).

The change of land use from forest to agricultural, residential and urban use has created a change in the O₃ precursor environment, which can be seen clearly between urban and rural areas (Kulkarni et al., 2016; Tu et al., 2007; Xie et al., 2016). This phenomenon usually leads to the formation of high concentrations of O₃ downwind and in suburban areas due to increased concentrations of O₃ precursors generating high concentrations of

O₃ (An et al., 2015; Latif et al., 2012). The high intensity of sunlight in tropical regions accelerates the process of O₃ formation (Ahamad et al., 2014; Awang et al., 2016a; Banan et al., 2013; Latif et al., 2012). Motor vehicles and industrial activities are the main sources of O₃ precursors such as NO_x and CO (Vingarzan, 2004; Xie et al., 2016). Areas with high volumes of vehicles have increased NO levels, which can then produce high concentrations of NO₂. The transport of NO₂ downwind will release O radicals which will then interact with O₂ to form O₃. At the same time, high concentrations of NO will effectively titrate O₃ in urban areas. Volatile organic compounds (VOCs) are other precursors of O₃ that can be emitted from anthropogenic as well as natural sources (Shao et al., 2016; Tie et al., 2006). Forest areas have been found to contribute to the high levels of VOCs; an example is isoprene which can transform to other organic peroxides and then contribute to enhanced concentrations of the O radical (Stavrakou et al., 2014; Taraborrelli et al., 2012). The combination of anthropogenic and biogenic O₃ precursor species usually leads to high concentrations of O₃ in the areas between highly populated urban centres and more forested or rural areas.

The island of Borneo is internationally recognised for its extensive, diverse and relatively untouched environment. The expansion of urban areas due to increasing population has changed the landscape of this island. However, agricultural activities are still the main economic generators for this area. Information on O₃ and precursor trends could indicate the potential need for mitigation measures. Studies by Sicard et al. (2013) and Sicard et al. (2016) for example, have analysed pollutant trends to evaluate effectiveness of O₃ control measures and determine more suitable standards for human health and environmental

protection. This study aims to determine the variations, particularly seasonal cycles and long term changes of surface O₃ in Borneo based on long term observational data provided by the Malaysian Department of Environment (DOE). The concentration of surface O₃ in relation to concentrations of O₃ precursors such as NO_x, CO and VOCs as well as other meteorological factors, such as wind speed and direction, UV radiation and temperature were analysed.

2. Methodology

2.1 Study area

Borneo is the largest island in Asia and home to one of the oldest rainforests in the world. The island is divided among three countries, Malaysia and Brunei in the north and Indonesia to the south. In the north, the East Malaysian states of Sabah, Sarawak and the federal territory of Labuan make up about 26% of the island. Kalimantan is located in the southern part of Borneo and belongs to Indonesia. Most major towns in Borneo are located in coastal areas due to the mountainous setting in the middle of the island. During the dry season, especially between January and March, and between July and September every year, biomass burning from peat swamp areas usually affects the air quality over Borneo.

This study analysed the variation of O₃ concentrations recorded at stations managed by Alam Sekitar Sdn Bhd (ASMA), a company which measures the air quality status on behalf of the Malaysian DOE in Sabah and Sarawak, Malaysian Borneo. Seven air quality stations in different areas around North Borneo had been chosen based on the availability of the O₃ air quality data during the study time period (Figure 1). Kuching station (S1) is

located at a medical store in Pending Industrial Estate, Kuching, Sarawak. This area is located on the outskirts of Kuching city. Sibu station (S2) is located at the Sibu Police Headquarters, close to the roadside in the city centre of Sibu. Both Kuching and Sibu stations are in busy areas where high volume of motor vehicles going in and out of the industrial areas, particularly during the rush hour. Bintulu station (S3) is located at the Bintulu Police Headquarters, on the outskirts of Bintulu city centre. The station is close to a less busy road next to a river, while to the north is an industrial area dominated by petrochemical industries (Liquid Natural Gas). Miri station (S4) is located in a school compound in the city centre of Miri near to a busy road. Miri is also located near to the border of Brunei. Both Miri and Brunei have extensive petroleum-related industries in the area. Labuan station (S5) is located in a housing area in centre of the Labuan Island. The station is located in the north of Labuan city. This station is expected to have less influence from motor vehicles compared to other stations. Kota Kinabalu station (S6) is located in a school compound in Putatan, a small town to the south of Kota Kinabalu city centre. The air quality is expected to be affected by the expansion of Kota Kinabalu city towards the south, including Putatan town. The number of motor vehicles during rush hour may influence the level of air pollutants at the station. Tawau station (S7) is located in a residential area of Tawau on the east coast of Sabah. The station located to the east of Tawau city centre which is one of the main towns on the east coast of Sabah. Detailed locations of the sampling points are presented in Supplementary 1.

[Figure 1]

[Supplementary 1]

2.2 Ozone and other parameter data collection

The air pollution data recorded at the ASMA stations between January 2002 and December 2013 includes the hourly average of surface O₃ concentrations, oxides of nitrogen (NO and NO₂), and carbon monoxide (CO). In addition to air pollutant parameters, the stations also recorded temperature, relative humidity, wind speed and wind direction. For Kuching station (S1), non-methane hydrocarbon (NmHC) and UV_b radiation were also recorded.

The surface O₃ concentrations at the ASMA air monitoring stations were measured using the UV Absorption O₃ Analyser (Teledyne Model 400A, USA). The analyser uses a system based on the Beer-Lambert law for measuring low ranges of O₃ in ambient air. The concentrations of NO and NO₂ were determined via chemiluminescence measurement, coupled with state-of-the-art microprocessor technology, used for monitoring high and medium levels of nitrogen oxides (Teledyne Models 200A, USA). Meanwhile, the concentrations CO and NmHC (representing total VOCs) were only determined at Kuching station (S1) and were measured using infrared energy absorption (Teledyne Model 300) and the field-proven Flame Ion Detector (FID) (Teledyne Model 4020, USA) respectively (Latif et al. 2012).

2.3 Data analysis

The daily maximum concentrations were used to compare the surface O₃ concentrations in different months and years. For the purpose of examining the relationship between O₃ concentrations and its precursors recorded at particular stations (NO, NO₂ and CO), hourly

data were used. The missing values for the data collected during the study period were between 6 to 12%. (Table 1). Missing values in the study period are not considered in the computation of statistics.

[Table 1]

2.4 Trajectories analysis

Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT version 4.9) developed by the National Oceanic and Atmospheric Administration (NOAA)'s Air Resource Laboratory (ARL) was employed to calculate 72 h back trajectories to determine origin of air mass arriving at the monitoring stations (Draxler et al., 2013). The height level selected for model calculation was 500 m to ensure that the trajectories started in the atmospheric boundary layer (Eva and Lambin, 1998). The meteorological drivers used to compute the trajectories from 2012 to 2013 were obtained from the National Centers for Environmental Prediction (NCEP) archive which is maintained by ARL. The one year period was selected as a representative year to examine the general transport pattern driven by the synoptic monsoon circulation and 2013 is also a year with reported haze occurrence. In order to compare the origin of air mass between two monsoon periods, clustering method in HYSPLIT model was also used to group the trajectories obtained based on curvature, length and transport characteristics.

3. Results and discussion

3.1 Descriptive analysis

The descriptive statistics of hourly surface O₃ concentrations from 2002 until 2013 are presented in Table 1. The hourly average concentrations of surface O₃ at the monitoring stations were recorded between 24 and 31 ppbv. Bintulu (S3) and Labuan (S5) stations

both recorded the highest average surface O₃ concentrations (31 ppbv). This was followed by Miri (S4), Sibul (S2), Kota Kinabalu (S6), Tawau (S7) and Kuching (S1). The median values were close to the average O₃ concentration (± 2 ppbv) while the standard deviation for the dataset was between 8 and 11 ppbv for all the stations. Although Labuan (S5) is the only monitoring station located at an altitude of higher than 10 m above sea level (ASL), its altitude is only 15 m ASL. Its average and median O₃ concentrations were similar to that of Bintulu station (S3) while its maximum O₃ concentration fell within the range of 61 – 92 ppbv recorded by the seven stations. There were significant differences ($p < 0.05$) in overall hourly surface O₃ concentrations between the surface O₃ recorded at the different monitoring stations. The different background atmospheric compositions at the urban and suburban stations, due to the different local emissions of O₃ precursors from motor vehicles and industrial activities, is thought to be the main reason for the different O₃ concentrations across all stations in long term hourly measurements (Banan et al., 2013).

Surface O₃ concentrations in Borneo are considered low compared to similar studies focusing on the Klang Valley, the busiest area located around Kuala Lumpur, the capital city of Malaysia (Ahamad et al., 2014; Banan et al., 2013; Latif et al., 2012). The stations located within and around Klang Valley frequently recorded maximum hourly concentrations that exceeded the limit of Malaysian Air Quality Standard of 100 ppbv. In Malaysian Borneo, the highest hourly concentration recorded was 92 ppbv at Bintulu station (S3). The concentrations of surface O₃ in Borneo were closer to the O₃ concentrations observed in two rural areas in Thailand (Inthanon and Srinakarin) by Pochanart et al. (2001) and eastern Thailand by Assareh et al. (2016). Higher surface O₃

concentrations were recorded in the densely populated residential-commercial area of Jakarta, Indonesia (Permadi and Kim Oanh, 2008). The lower concentrations recorded in these stations in Malaysian Borneo are expected due to the lower concentrations of surface O_3 precursors emitted from motor vehicles and industrial activities compared to the stations in the big cities such as Kuala Lumpur and Jakarta. The lower emissions of O_3 precursors will reduce the ability of surface O_3 to be produced around the downwind areas. Nevertheless, the stations in Borneo recorded surface O_3 concentrations higher than the surface O_3 recorded at the Malaysian Department of Environment (DOE) background stations in Jerantut, Pahang. The average concentration recorded here is around 13 ppbv for surface O_3 , as shown by Latif et al. (2012), Banan et al. (2013) and Latif et al. (2014).

3.2 Diurnal pattern of surface O_3 concentrations

The diurnal pattern of air pollutants recorded in this study, as well as the UV_b recorded at S1, showed similar patterns to previous studies (Ahamad et al., 2014; Awang et al., 2016a; Banan et al., 2013; Latif et al., 2012). O_3 showed a single peak during daylight hours while the precursor concentrations showed bimodality which is attributed to traffic patterns (Figure 2). The concentration of surface O_3 was highest at midday at S1, when the intensity of UV_b was at the highest level. A high level of sunlight will encourage the formation of the O radical, which is responsible for the formation of O_3 from its precursors such as NO_x and VOCs (Awang et al., 2016b; Xie et al., 2016; Zhang et al., 2014). The concentrations of O_3 precursors such as CO, NO, NO_2 and NmHC (recorded at S1 only) recorded at their highest levels during rush hour between 7:00 and 9:00 am. At midday, the concentrations of these precursors were very low, due to their transformation to other

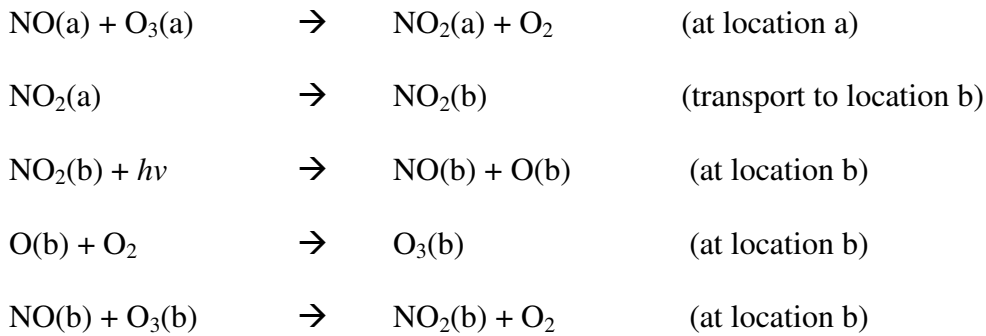
secondary species of pollutants (including O_3) as well the convective transfer of pollutants to higher altitudes. The concentrations of the surface O_3 precursors peaked again in the evening when people travel back home from work. The concentrations of O_3 precursors showed a less steep drop at night due to the higher atmospheric stability during the night and the absence of sunlight, reducing the scavenging reaction of photochemical oxidants (Awang et al., 2015).

[Figure 2]

3.3 Ratio between NO_2 and NO

The ratio of NO_2/NO has been used as an indicator to determine the shift in equilibrium towards the formation of O_3 from NO_2 . When the ratio is high, O_3 formation is typically favoured as NO can both contribute to the formation of NO_2 and the destruction of O_3 (Ismail et al., 2016; Xie et al., 2016). Table 2 shows the ratio of NO_2/NO during the period when the O_3 peak is typically observed. The ratio is calculated based on total averaged NO and NO_2 concentrations. The ratios were recorded at higher values at both S3 and S4 while all other stations recorded a consistent ratio close to one. These two stations are located in the areas associated with petrochemical industries which have the potential to release high amount of NO_x and VOCs. The other stations, located in the city centre and suburban as well industrial areas, did not indicate any excess of NO_2 over NO . This result is similar to the study by Latif et al. (2012) in Peninsular Malaysia, which showed that stations with high NO_2/NO ratios, especially in suburban areas, recorded high concentrations of O_3 compared to stations in the city centre.

The NO₂/NO ratios indicate that there is potential for high O₃ generation due to the formation of NO₂ from NO at two of the stations (S3 and S4). During the day, NO₂ can be quickly photo-dissociated with subsequent O₃ formation. However, NO₂ release at night and in the early morning can influence nearby areas and produce high amounts of O₃ at midday as suggested by Geng et al. (2008). Overall interactions of NO_x at the sampling station (b) from its potential source areas (a) was explained by Geng et al. (2008) as below. Titration process at (b) will reduce the concentration of O₃ at the sampling station.



[Table 2]

3.4 Weekday and weekend effect

The normalised values of surface O₃ concentration based on the difference between the average concentration of daily O₃ and weekly average during weekdays and weekends at all stations are shown in Figure 3. The results show that the concentration of surface O₃ was recorded at higher concentrations during the weekend compared to weekdays. The results also show decreasing concentrations of O₃ precursors (NO_x and CO) from weekdays to weekends. The pattern of higher O₃ at weekends compared to weekdays has been recorded in other studies in Malaysia such as Latif et al. (2014) and Banan et al

(2013). This pattern is usually seen in the suburban and rural areas which have significantly different numbers of motor vehicles during weekdays compared to weekends.

[Figure 3]

During weekdays, the number of motor vehicles on the road will increase compared to weekends due to the movement of people going to work and children going to school. The low concentration of NO emitted by motor vehicles as well as industrial activities, especially in the middle of the day, during the weekend will reduce the capability of NO titrate O₃ in ambient air (Alghamdi et al., 2014; An et al., 2015; Wang et al., 2014). The concentration of O₃ therefore will be higher during the weekend compared to weekdays. Titration of O₃ by NO that is emitted by anthropogenic sources contributes to the weekend effect. The weekend effect seen at the stations in this study indicate that the profile of these stations are closer to suburban than urban due to the difference in the concentrations of O₃ and O₃ precursors between weekdays and weekends. Although some of these stations are urban-industrial stations due to the activities carried out, the population density is less than highly-urbanised city centres.

3.5 Monthly variation of O₃ concentration

The monthly average daily maximum concentrations of surface O₃ at all monitoring stations, except for station S7, were highest in August (Figure 4). This month can be considered the dry month in the Southeast Asian maritime continental area compared to other months of the year. Haze episodes usually occur due to biomass burning, particularly from Kalimantan, Indonesia, and will add to the amount of O₃ precursors at the monitoring

stations along the west coast and north of Borneo due to the wind direction of the southwest monsoon.

[Figure 4]

Based on the clustered HYSPLIT model provided in Supplementary 2, wind movement during the southwest monsoon originates from the southern part of Borneo. This wind influences the pollutant concentrations, which are dominated by biomass burning as well as emissions from urban and industrial areas along the west coast of Borneo. This wind movement is expected to contribute high amounts of surface O_3 compared to the northeast monsoon. It should be pointed out that the local scale wind system such as the land-sea breeze may also modulate the concentration of pollutant near the coastal regions. However, the HYSPLIT model was driven by the large scale meteorological field and hence does not account for such effect in the calculation of the trajectories. According to He et al. (2010) haze episodes can contribute to the amount of polar organic compounds such as phthalic acid and cis-pinonic acid. These two acids showed strong linear relationships with maximum daily O_3 concentrations throughout the entire sampling period during haze episodes occurring in Singapore in 2013. The high intensity of UV radiation during the dry period when there is less cloud cover may also contribute to the formation of surface O_3 (Awang et al., 2016a; Geng et al., 2008; Xie et al., 2016). The wet conditions at the turn of the year (November to January) usually reduce the amount of surface O_3 precursors in these stations due to washout of pollutants and high cloud cover.

[Supplementary 2]

Conversely Tawau station (S7) recorded the highest O₃ concentrations between November and February, which is during the northeast monsoon. A likely reason for the difference is the location of the station. Unlike S1 – S6, which are located on the west coast of Borneo, Tawau (S7) is located on the east coast which is influenced by different wind directions. The wind during the northeast monsoon (Supplementary 3) is arriving from the sea to most of the stations on the west coast of Borneo. The northeast monsoon is expected to bring more surface O₃ precursors because the wind during the northeast monsoon moves towards Tawau via land compared to the southwest monsoon, which brings in air masses over the sea.

[Supplementary 3]

3.6 Yearly pattern of O₃ concentration

The time series of the annual mean of maximum concentrations of surface O₃ are shown in Figure 5. The detailed information on trends based on O₃ yearly maximum concentrations and yearly average O₃ concentration, as well as O₃ precursor (NO, NO₂, CO) yearly average concentrations, are presented in Table 3. A majority of stations (except S1 and S6) have recorded increasing averaged maximum concentrations of surface O₃ over the analysed years. In particular, O₃ recorded at stations S3 and S5 have increased by 0.26 and 0.28 ppbv/year, respectively, which are larger than their respectively natural variability. From these two stations, only S3 shows a significant increased trend ($p < 0.05$). Stations S1 and S6, which are located near to congested city centres (Kuching and Kota Kinabalu), showed a decreasing trend. Generally, the linear trends of the maximum O₃ corresponds to the changes of the annual mean of the O₃ concentration despite different increasing rates,

except at S1, where the average O₃ concentration increased slightly (0.09 ppbv/year) when the maximum O₃ decreases (-0.18 ppbv/year). This suggests that different processes modulate the changes at different part of the O₃ distribution in these stations for the time-scale considered. In particular, the yearly maximum value is a representative of midday maximum O₃ concentration, which can be modulated by regionally generated photochemical episodes and meteorological factors. The annual averaged, on the other hand is based on total 24-h averaged values and can be mostly influenced by the magnitude of local emission and processes.

[Figure 5]

Overall, yearly variations of surface O₃ concentrations are expected to be influenced by local and transboundary O₃ precursors as well as variations in meteorological factors. Increasing yearly trends of NO₂ concentrations (S1, S2, S3 and S6) potentially increase the concentration of O₃ in the sampling locations. Nevertheless, increasing trends of NO such as at S6 (0.22 ppbv/year) and S1 (0.08 ppbv/year) may also reduce the O₃ due to the titration process. The high increasing trends of O₃ at S3 can be associated with increasing trends of NO₂ and decreasing trends of NO. On the other hand, a high reduction of NO compared to NO₂ may influence the increasing trend of O₃ at S5.

There was no clear indication of the influence of yearly CO trends on the concentrations of yearly surface O₃. Decreasing trends of CO, especially at S1 (-30.37 ppbv/year), may suggest the reduction of incomplete combustion from vehicles and industrial processes. The reduction of CO has led to increasing trends of NO₂, which is expected to come from

complete combustion processes. Nevertheless, due to increasing trends of NO, the concentrations of O₃ were not found to be influenced by high concentrations of NO₂ at this station. The trend of CO from S4 to S7, which correspond to the fluctuation of NO, was expected to be influenced by motor vehicle emissions. S3 showed unique CO, NO and NO₂ trends, where an increasing trend of CO was not followed by an increasing trend of NO. CO at this station may have the capacity to increase the concentration of CO in the surrounding areas.

3.7 Bivariate polar plot

Hourly O₃ and NO₂ measurements are shown on bivariate polar plots to illustrate how the pollutant variations are affected by wind speed and wind direction. Polar plots have been applied to discriminate source types, provide additional information on complex wind-pollutant interactions and vertical updrafts (Carslaw et al., 2006; Tomlin et al., 2009; Westmoreland et al., 2007). General features observed are that all the seven stations show evidence of increases in O₃ concentration as the wind speed increases (Figure 6). The low concentration of NO₂ at high wind speed is expected as NO₂ is dispersed in the presence of high temperatures and sunlight, with NO₂ releasing an O radical that contributes to the formation of surface O₃. The higher concentrations of NO₂ at low wind speeds are the result of more stable atmospheric conditions and reduced advection that exist under low wind speed conditions. Despite the influence of urbanisation and industrial emissions, the sea breeze appears to play an important role in the distribution of the precursors of O₃ (Latif et al., 2012). As mentioned by Physick and Abbs (1992), pollutants can be transported many kilometres inland by the sea breeze circulation.

[Figure 6]

S3, which has the highest hourly surface O₃ concentrations, appears to show an obvious maximum concentration when the wind is from the north and northwest. S4 also showed high O₃ concentrations when the wind originated from the northwest. These features are observed with wind speeds between 5 m/s to more than 30 m/s for both stations (Figure 6). Bintulu (S3) and Miri (S4) stations are located close to the large petrochemical and natural gas industries in the northwest. S5 is an island where the sea breeze blows from almost all directions especially from the northeast, northwest and southwest. S1 and S6 stations did not show a clear pattern of concentration with wind direction. The high O₃ concentrations were more scattered which indicates that the pollutant sources were the surrounding industrial activities and also long-range transport of air pollutants. An increase in O₃ in Sibul (S2) was observed when the wind originated from the southwest, and for Tawau (S7) the O₃ concentrations were apparent when the wind was from the south and southeast. Concentrations of O₃ at these two stations are expected to originate from O₃ precursors that are at high levels due to urbanisation activities surrounding the sampling stations. Nevertheless, O₃ and its precursors coming from transboundary emissions such as biomass burning from the south and southwest directions will be important.

3.8 Relationship between O₃ and its precursors and meteorological factors

Density plots were drawn to determine the relative influence of precursor concentrations and meteorological factors on surface O₃ concentrations recorded at all stations (Figure 7).

The plot of O_3 against NO shows that higher O_3 concentrations are mostly observed at low NO concentrations (< 10 ppbv). At higher NO concentrations, titration of O_3 is expected to dominate the interaction between O_3 and NO. The density and concentration of O_3 were higher for NO_2 (10 - 20 ppbv) compared to NO. However, when the NO_2 concentration was > 40 ppbv, O_3 concentrations were much lower than the maximum concentration observed at < 20 ppbv NO_2 . The negative relationship between O_3 and NO_2 is expected as NO_2 is a precursor to O_3 . The plot of O_3 against CO shows a similar profile to NO_2 with the exception of higher O_3 concentrations at high CO concentrations (> 4000 ppbv). The highest O_3 concentrations were recorded when the CO concentration was around 500 to 1500 ppbv. Temperature shows a positive relationship with O_3 until about $35^\circ C$, after which O_3 concentration does not show an apparent increase despite increasing temperatures. In the plot of O_3 against wind speed, the highest density of O_3 concentrations are observed at speeds of < 5 m/s. O_3 concentrations were recorded at the highest values when the wind speed was between 5 to 15 m/s. Due to the high humidity in this region throughout the year, the highest density of O_3 is observed around relative humidity of 90%. However, O_3 concentrations above 60 ppbv are only observed when relative humidity is less than 80%. A reduction in humidity is typically accompanied by higher temperatures and hence the increase in O_3 in Malaysia (Latif et al., 2014; Latif et al., 2012). Also, water molecules can react with the photolysis product of O_3 and form hydroxyl radicals which subsequently contribute to O_3 formation in the presence of other precursors (Seinfeld and Pandis, 2006).

[Figure 7]

4. Conclusion

The results of the study show that the concentration of surface O_3 in the northern part of Borneo (Malaysian region) are still lower than the maximum permissible value of 100 ppbv suggested by the Malaysian Air Quality Standard for 2015 (Interim period - 1). The concentration of surface O_3 is expected to be influenced by the amount of NO_x , particularly from motor vehicles and the petrochemical industry. Stations located near to petrochemical sources such as Bintulu (S3) and Miri (S4) reported higher concentrations of O_3 than other stations. Higher ratios of NO_2/NO at these stations showed that NO_2 is a potential major contributor to the amount of O_3 . Titration processes due to the concentration of NO from motor vehicles reduce the concentration of surface O_3 , especially at the stations near to the city centre such as Kuching (S1) and Kota Kinabalu (S6) and especially during weekdays. At the weekend higher concentrations of O_3 are observed. Meteorological factors, especially temperature, UV radiation and wind speed, are found to influence the concentration of surface O_3 in Borneo. Local and regional winds are also found to influence the concentration of surface O_3 as shown at the Labuan station (local effect). High concentrations of O_3 during the southwest monsoon may be due to biomass burning (transboundary effect) but this needs more detailed study and investigation. Majority of the stations (except S1 and S6) have recorded increasing averaged maximum concentrations of surface O_3 over the analysed years. The increasing significant trends may due to the combination of increasing trends of NO_2 and decreasing trends of NO . The density plots shows that the concentration of surface O_3 was highest when the concentration of its precursors were at a certain range ($NO < 10$ ppb; NO_2 : 10-20 ppbv; and CO : 500 – 1500 ppbv). Particular range of wind speed (5 to 15 m/s),

temperature ($< 35^{\circ}\text{C}$) and humidity ($< 80\%$) were also found to be conducive for increasing surface O_3 .

This study suggests that more detailed studies of surface O_3 and its precursors in Borneo are required, especially the relation of surface O_3 to other O_3 precursors such as VOCs from various anthropogenic and natural sources. To maintain the concentration of O_3 below the maximum value, especially in the middle of the day, the concentrations of O_3 precursors need to be monitored and controlled. The emission of NO_x with high concentrations of VOCs will increase the concentration of surface O_3 in the study areas, especially in areas located close to petrochemical industries. There is also a need to study the effects of the long-range transport of surface O_3 and its precursors, such as emission from biomass burning in other parts of Southeast Asia, especially Kalimantan. This study indicates that the biomass burning, which occurs from June to September every year, can contribute to the high concentration of surface O_3 in Malaysian Borneo.

ACKNOWLEDGEMENTS

The authors would like to thank Universiti Kebangsaan Malaysia for the Research University Grants (DIP-2014-005 and AP-2015-010). Special thanks to Malaysian Department of Environment (DOE) for providing air quality and meteorological data for the process of conducting our research. We gratefully acknowledge the NOAA Air Resources Laboratory (ARL) for the provision of the HYSPLIT transport and dispersion model and READY website (<http://www.arl.noaa.gov/ready.html>). We also gratefully thank Natural Environment Research Council (NERC) *openair* project

(<http://www.openair-project.org/>). Special thanks to Dr Rose Norman for proofreading this manuscript.

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Table's Caption

Table 1: Locations of air quality station around the Borneo Island (Sabah and Sarawak, Malaysia) and descriptive data for hourly surface O₃ hourly concentration from 2002 until 2013

Table 2: Ratio of NO₂/NO concentrations between 12 pm and 4 pm (2002 – 2013)

Table 3: Yearly averaged O₃ trend for hourly maximum O₃, and yearly averaged O₃ trend for hourly average O₃ and its precursors (2002 – 2013). Values in bold are significant at $p < 0.05$.

Figure's Caption

Figure 1: Location of seven selected air quality monitoring stations

Figure 2: The diurnal pattern of surface O₃ and its precursors at all monitoring stations. Only station S1 recorded concentration of NMHC and UV_b

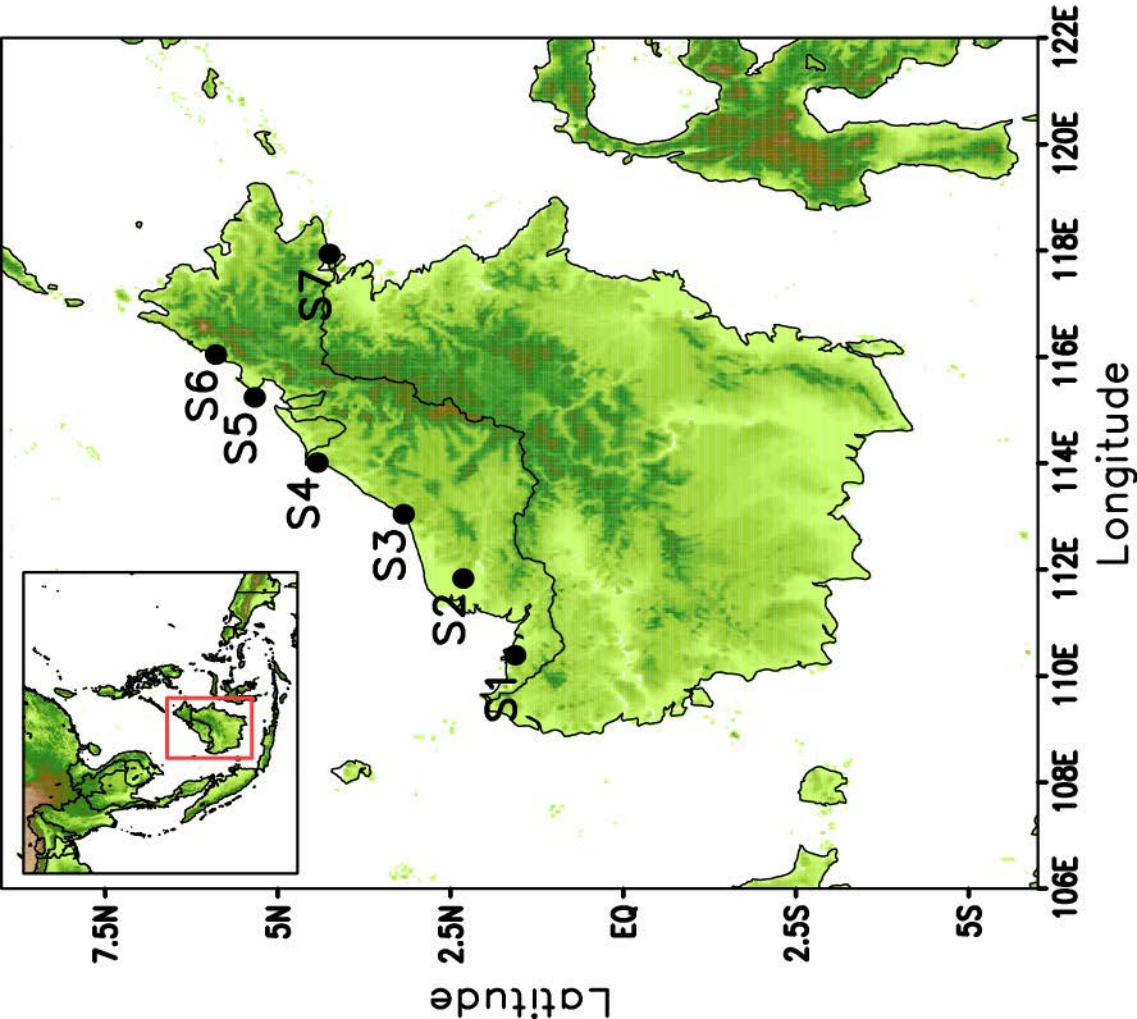
Figure 3: Concentration of surface O₃ and its precursors (CO and NO_x) during week days and weekend

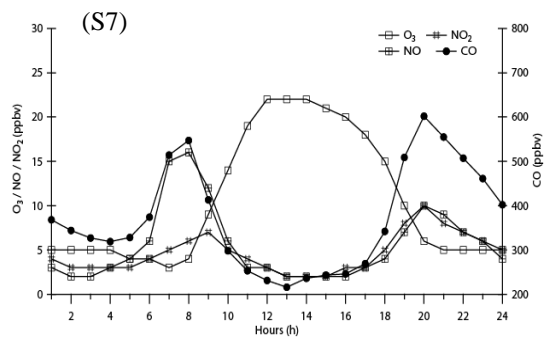
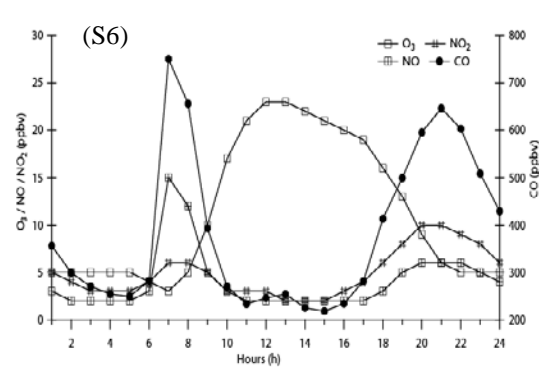
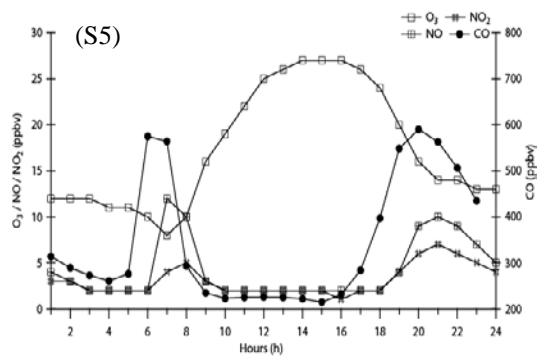
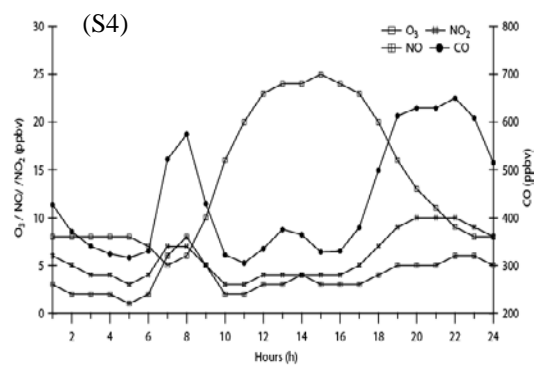
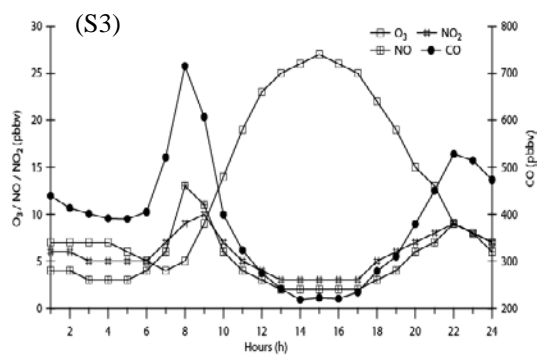
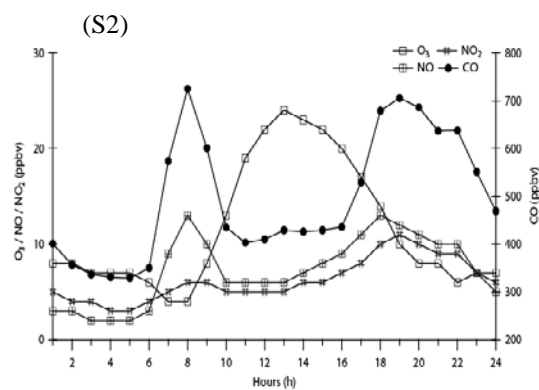
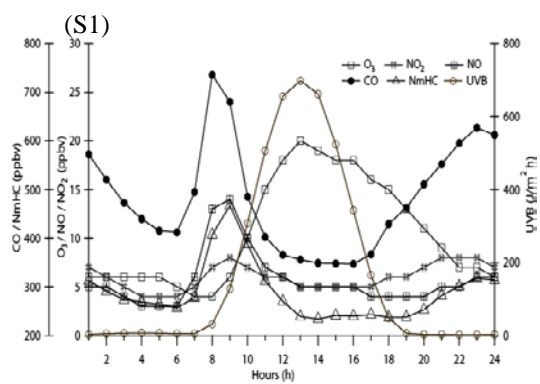
Figure 4: Monthly surface O₃ daily maximum recorded at the monitoring stations

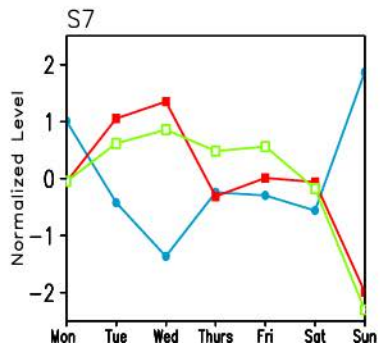
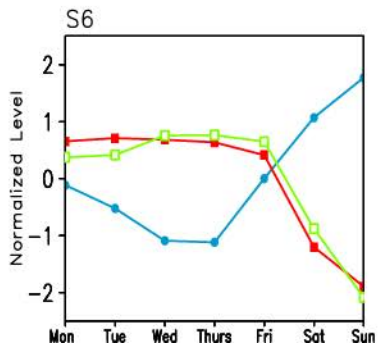
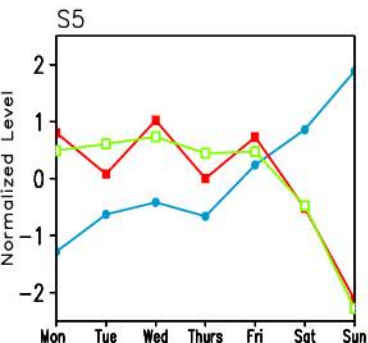
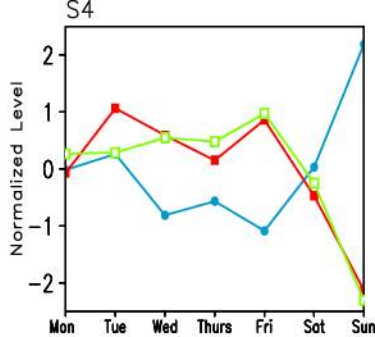
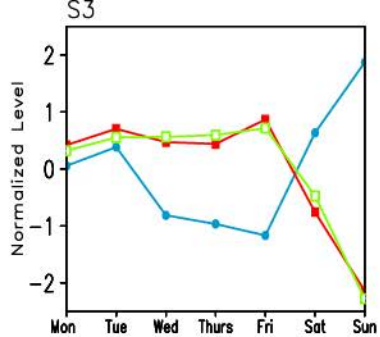
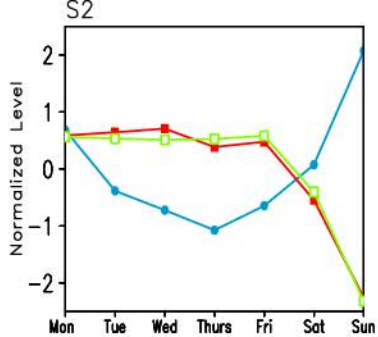
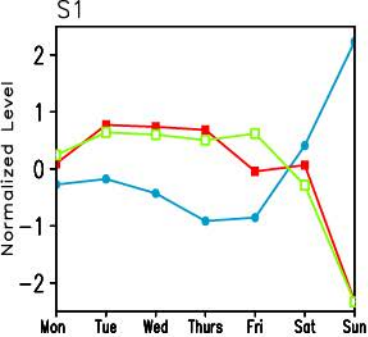
Figure 5: Yearly mean of daily maximum O₃ concentration recorded at the monitoring stations

Figure 6: Bivariate plot of O₃ (ppbv) at each station. The centre of each plot represents wind speed, starting from zero and increasing radially outward. The O₃ concentration is coded using a colour scale.

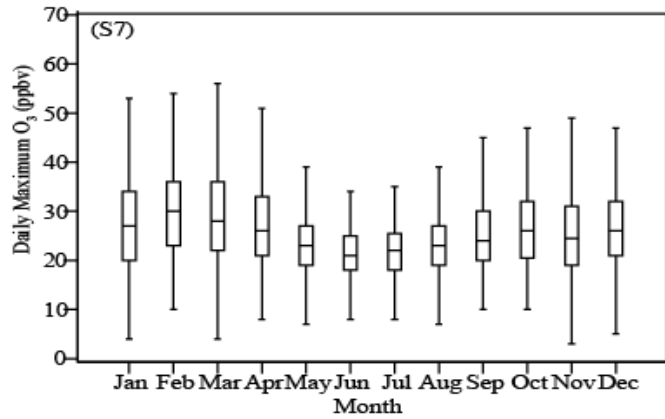
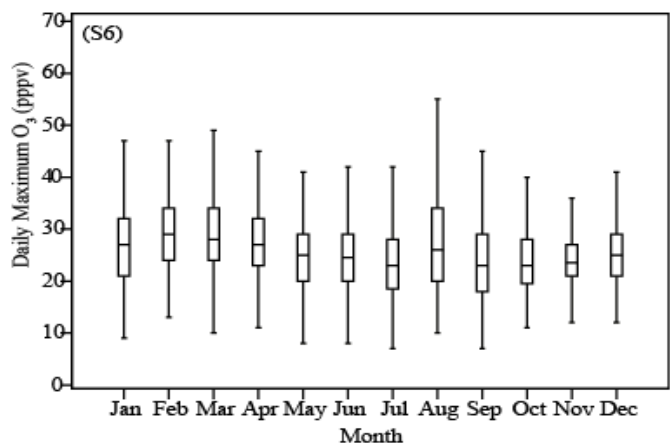
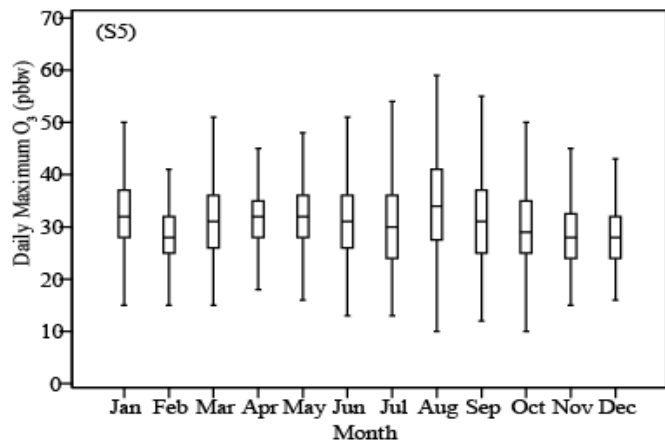
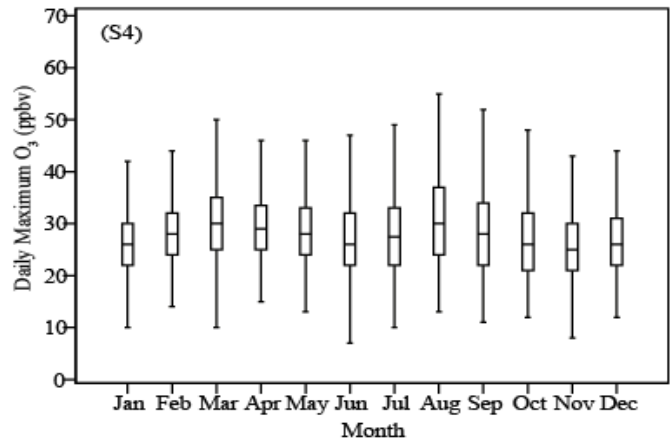
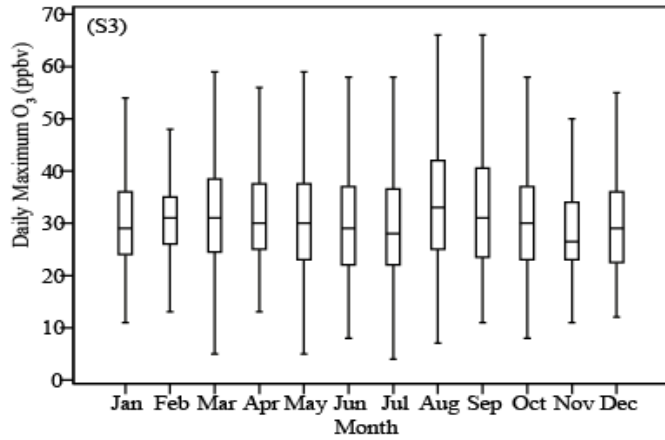
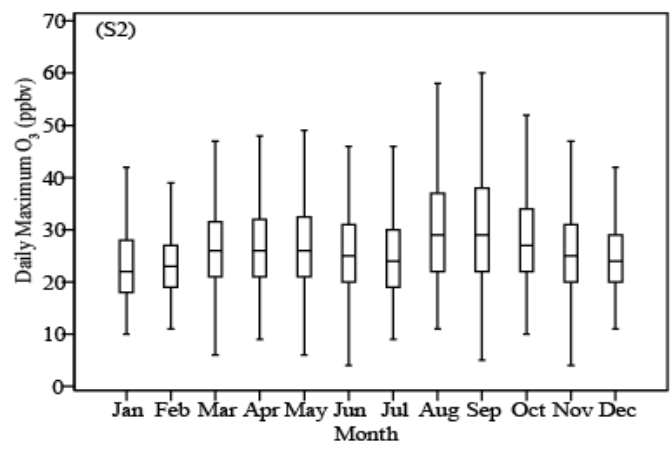
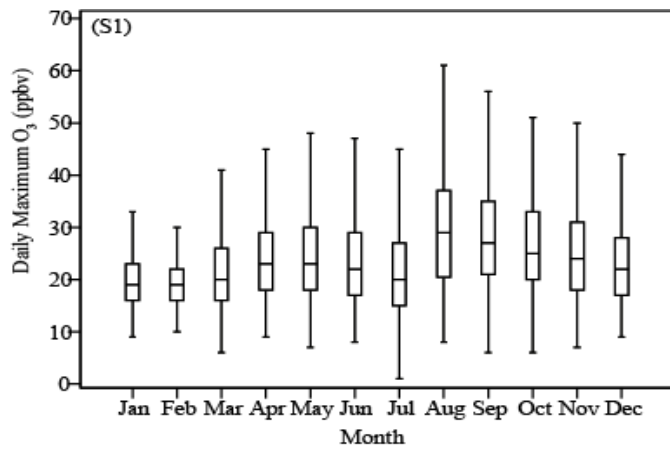
Figure 7: Scatter plot between O_3 and its precursors (CO, NO and NO_2); and between O_3 and meteorological factors (Temperature, Humidity and Wind Speed)

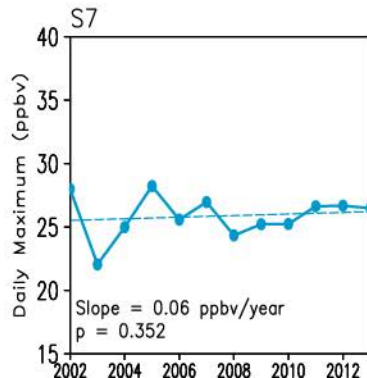
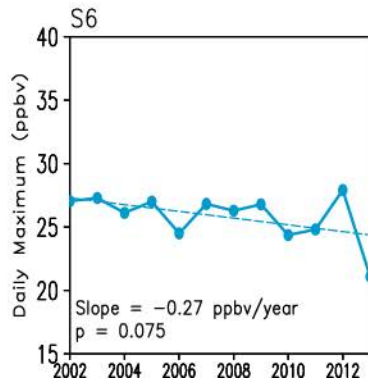
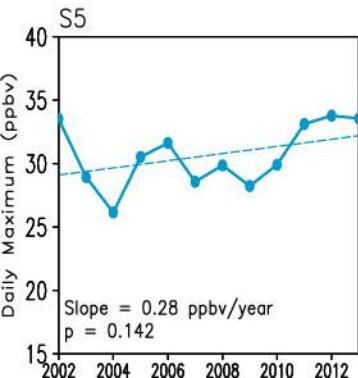
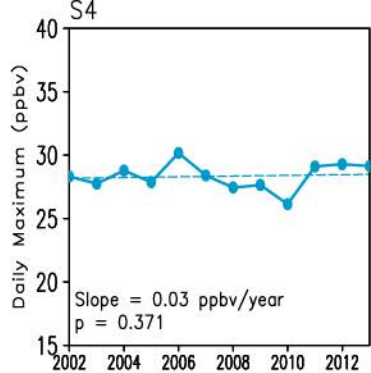
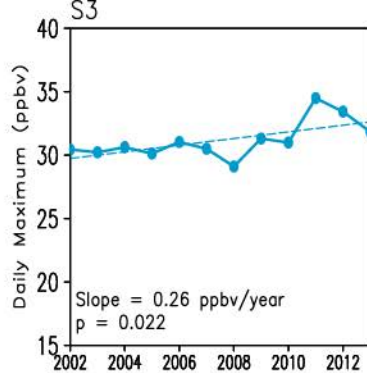
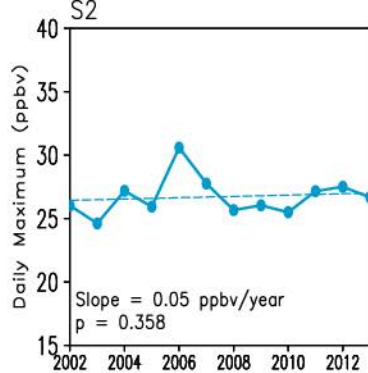
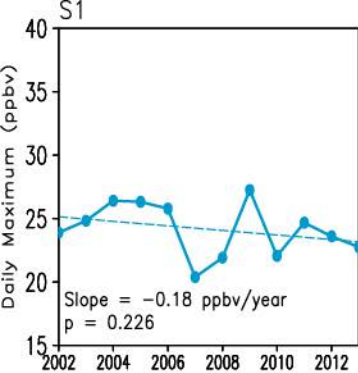


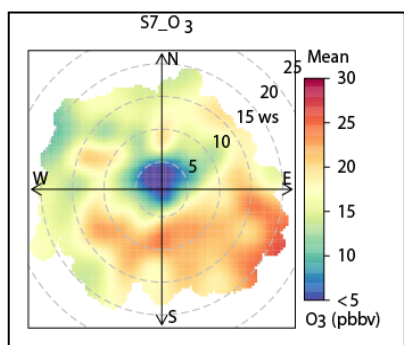




—●— O3
—■— CO
—□— NOx







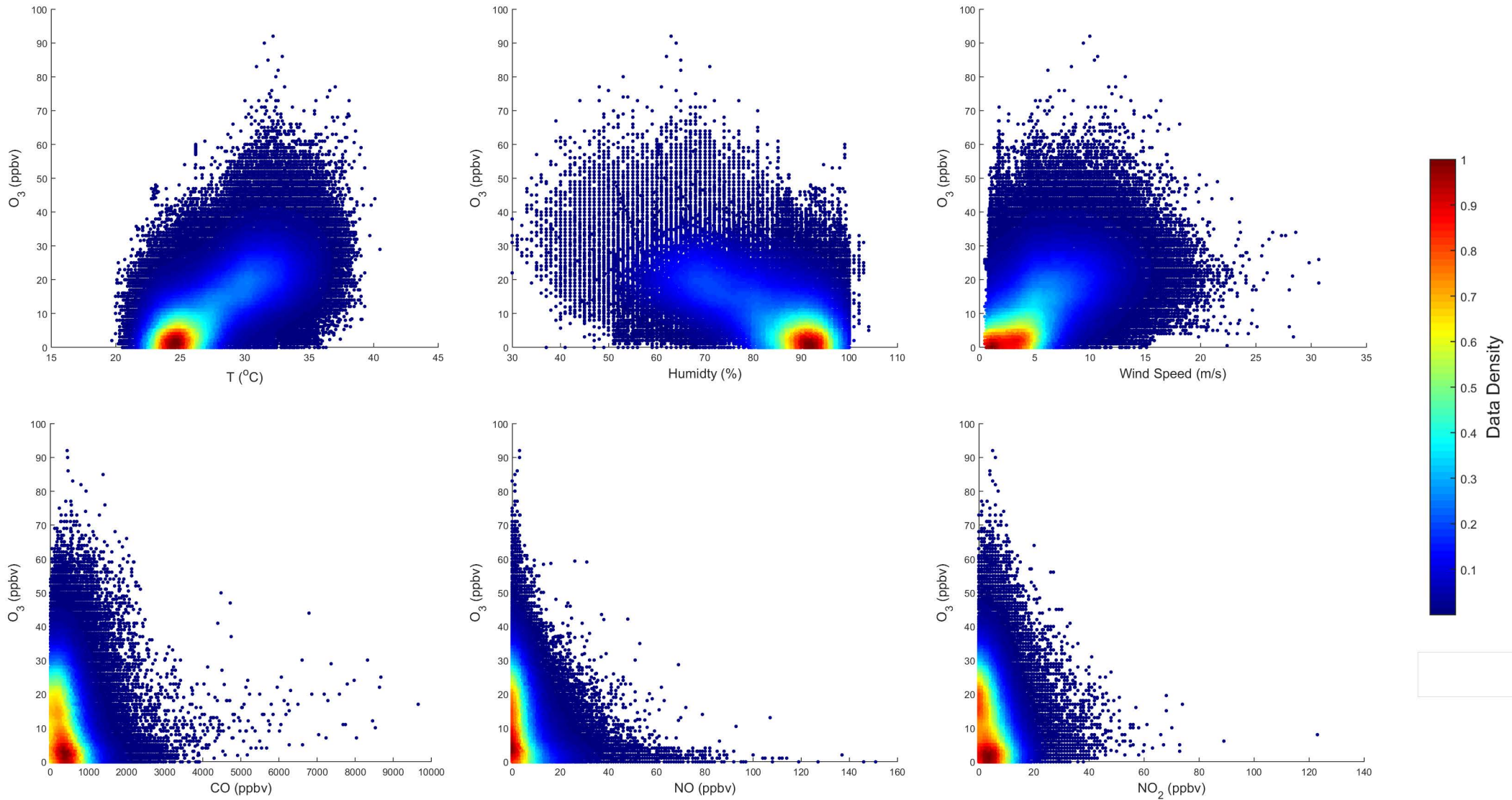


Table 1: Locations of air quality station around the Borneo Island (Sabah and Sarawak, Malaysia) and descriptive data for hourly surface O₃ hourly concentration from 2002 until 2013

Location	Coordinate		Altitude ^a (m)	Missing value ^b (%)	Hourly Ozone Concentration (ppbv)			
	Latitude	Longitude			Average	Standard Deviation	Median	Maximum
Kuching (S1)	N01° 33.734	E110° 23.329	7	12	24	10	22	82
Sibu (S2)	N02° 18.856	E111° 49.906	9	9	27	9	25	67
Bintulu(S3)	N03° 10.587	E113° 02.433	8	10	31	11	30	92
Miri (S4)	N04° 25.456	E114° 00.731	6	8	28	8	28	80
Labuan (S5)	N05° 19.980	E115° 14.315	15	10	31	8	30	77
Kota Kinabalu (S6)	N05° 53.623	E116° 02.596	8	11	26	8	25	61
Tawau (S7)	N04° 15.016	E117° 56.166	7	14	26	9	25	69

Note: ^a Altitude base on above sea level (ASL)

^b Missing value based on data collection from starting date

Table 2: Ratio of NO₂/NO concentrations between 12 pm and 4 pm (2002 – 2013)

Station	12 pm	1 pm	2 pm	3 pm	4 pm
S1	0.90	0.97	0.98	1.06	1.19
S2	0.94	0.91	0.84	0.82	0.80
S3	3.21	4.70	5.42	5.36	5.67
S4	4.07	3.49	3.44	4.53	4.77
S5	0.98	0.87	0.81	0.84	0.75
S6	1.19	1.07	1.11	1.19	1.44
S7	1.05	1.04	1.03	1.06	1.10

Table 3: Yearly averaged O₃ trend for hourly maximum O₃, and yearly averaged O₃ trend for hourly average O₃ and its precursors (2002 – 2013). Values in bold are significant at $p < 0.05$.

Station	Yearly Maximum trend (ppbv/year)	Yearly Average trend (ppbv/year)			
	O ₃	O ₃	NO	NO ₂	CO
S1	-0.18	0.09	0.03	0.31	-30.37
S2	0.05	0.13	-0.20	0.30	-21.75
S3	0.26	0.14	-0.07	0.08	2.87
S4	0.03	0.13	-0.19	-0.05	-19.79
S5	0.28	0.16	-0.24	-0.03	-11.57
S6	-0.27	-0.10	0.22	0.27	1.29
S7	0.06	0.08	0.39	0.28	11.18